

REMARKS

The specification has been objected to, claims 15-16 have been rejected under 35 U.S.C. § 112, and claims 12-16 have been rejected under 35 U.S.C. §103(a).

An new Abstract has been provided to overcome the objection to the specification in Item 4 of the Office Action.

Claim 15 has proper antecedent basis. Claim 16 has been amended to overcome the antecedent basis rejection in Item 5 of the Office Action.

New claims 21-23 have been added to further define the invention of claim 12, and have a basis at paragraph [0015].

Claims 12-16 have been rejected under 35 U.S.C. § 103(a) in view of U.S. Patent No. 6,087,032 to Yoshitake *et al.* ("Yoshitake") and US 2002/0096279 to Kinsley Jr. ("Kinsley") and U.S. Patent No. 5,207,826 to Westland *et al.* ("Westland").

Yoshitake was cited as disclosing a fuel cell comprising an electrolyte membrane, a fuel electrode and an air electrode. The Office Action concedes that Yoshitake does not disclose specific anode or cathode material. It should be further noted that Yoshitake does not disclose the use of bacterial cellulose in an electrolyte membrane.

Kinsley was cited as teaching the use of bacterial cellulose for fuel cell electrode purposes. However, upon further review it can be seen that Kinsley does not disclose an electrode comprising bacterial cellulose. Attention is first directed to paragraph [0023] where it states that metal fibers may be dispersed in a slurry composed only of water and a high surface area material like bacterial cellulose. Then, at paragraph [0026] it states that "once the metal fibers have been dispersed in the aqueous

dispensing fluid, the dispensing fluid is then applied to a screen as is conventional in papermaking process,” and “the aqueous dispensing fluid is then removed in order to form the metal fiber sheet.” Thereafter, at paragraph [0031] it states that

The final step is a sintering step which can be conducted at optimum temperatures in an inert or reducing atmosphere. The sintering step introduces a strength to the metal fiber paper, as well as burns off the various organics contained in the metal fiber paper. The sintering step generally involves heating the paper at a temperature of from 1500-1200°F. for a time necessary to burn off the organics. (Underlining added.)

At paragraph [0037], it notes that the resulting metal fiber sheet can be used as a battery electrode.

These paragraphs from Kinsley clearly show that the electrode described in Kinsley does not contain bacterial cellulose as recited claim 12. The bacterial cellulose is used in Kinsley merely as a dispersing agent, and the sintering step in Kinsley will necessarily burn off the cellulose. Hawley's Condensed Chemical Dictionary lists the ignition point of cellulose as approximately 450°F and therefore, given the sintering temperature listed in Kinsley (1200-1500°F), no cellulose will remain in the resulting electrode. Thus, it is respectfully submitted that Kinsley fails to teach an electrode comprising bacterial cellulose.

Westland was cited in the Office Action as teaching that bacterial cellulose can be used as membranes and/or specialty components for fuel cells and/or materials having special electronic properties. In particular, the Office Action makes reference to column 2, lines 56-69 of Westland where Westland references the teachings of WO 89/12107 when describing various uses of bacterial cellulose.

Applicant was not aware of WO 89/12107 until Westland was cited in the Office Action and therefore, a copy of WO 89/12107 has now been reviewed. The Applicant encloses a Form PTO-1449 with this document listed and respectfully requests that WO 89/12107 be considered by the Examiner, be made of record in the present application and that an initialed copy of the Form PTO-1449 be returned in accordance with MPEP § 609.

Upon review of WO 89/12107, Westland appears to be making reference to page 3, line 28 to page 4, line 2 of WO 89/12107 where some uses of bacterial cellulose are broadly described. However, the mention of "membranes" in WO 89/12107 says nothing about fuel cell components. It is describing filtration and separation media such as membranes for water purification (see page 3, lines 28-31 of WO 89/12107).

WO 89/12107 does mention "a specialty carrier, such as for battery fluid and fuel cells" at page 3, lines 32-33. However, "a specialty carrier" could mean anything and in fact, page 17, lines 33-36 of WO 89/12107 describe the specific use of bacterial cellulose contemplated in WO 89/12107 as "gels useful in fuel cell and battery structure articles, where the electrolyte comprises the liquid phase of the microbial cellulose gel." Thus, the "specialty carrier, such as for battery fluid and fuel cells" described in WO 89/12107 and made referenced to by Westland is an electrolyte membrane where the electrolyte comprises the liquid phase of the microbial cellulose gel. Nothing in WO 89/12107 or Westland describes the use of bacterial cellulose in a fuel cell anode or a fuel cell cathode as specifically recited in claim 12.

WO 89/12107 also mentions "materials having special electronic effects produced by coating the individual microbial-produced, cellulose, microfibrils with

appropriate components, such 'as metals by vapor deposition or epitaxial growth" at page 3, line 34 to page 4, line 2. However, what is being referred to is the production of conducting, semiconducting or superconducting dry ribbons or thin films of metal coated cellulose (see page 11, lines 25-33 and page 14, lines 13-19 of WO 89/12107). There is no mention of the use of bacterial cellulose in a fuel cell anode or a fuel cell cathode as specifically recited in claim 12.

There are further fundamental differences between the present invention and the teachings of Yoshitake, Kinsley and Westland. The method used for formation of a membrane or sheet of cellulose in these patents is the same as that used for casting sheets of paper in the paper and pulp industries, that is, a suspension or slurry of mechanically disrupted cellulose fibers that are obtained from a natural source, either plant-derived or bacterial, is laid or rolled out and dried. The method of the present application uses the sponge-like bacterial cellulose pellicule without disruption of its natural structure and microarchitecture. The bacterial cellulose described by Westland is produced in agitated culture from a strain that produces cellulose in the form of loose pellets or pieces, instead of a contiguous, sponge-like gel pellicule, such as those employed by the present inventors. These hydrated pellicules can be treated with different chemicals to form metallized electrode materials or proton-conductive materials, and then assembled by serially drying one onto the other. A stable membrane sandwich is formed by hydrogen bonds between the hydroxyl groups of the cellulose chains.

The method used Yoshitake, Kinsley and Westland for incorporation of metal particles employ one of two processes: (1) preformed metal particles that are then

entrapped in the cellulose matrix during the sheet casting process; or (2) metal particles that deposited or epitaxially grown on the surfaces of the cellulose by vaporization of the metal or metals in question in special chambers. In our patent application, the preferred embodiment is the deposition of metal particles from the corresponding metal salts in aqueous solution that are infused into the natural cellulose structure. Particle formation is then initiated by reduction of hexachloropalladate by the reducing ends of the cellulose chains. Cellulose is a polymer of glucose, and the reducing ends have free aldehyde groups as does glucose in solution. An alternative method is the infusion of another chemical that can reduce the metal salts inside the cellulose matrix. The inventors have tunneling electron microscopy images of the palladium particles formed by this method that show size (5-20 nm) and crystallinity of the palladium particles.

In summary, (1) Yoshitake does not disclose the use of bacterial cellulose in an electrolyte membrane or a fuel cell anode or a fuel cell cathode as recited in claim 12; (2) the electrode described in Kinsley does not contain bacterial cellulose as recited claim 12; and (3) Westland, and WO 89/12107 which is referenced in Westland, merely describe a "specialty carrier, such as for battery fluid and fuel cells" which turns out to be an electrolyte membrane where the electrolyte comprises the liquid phase of the microbial cellulose gel and therefore, nothing in WO 89/12107 or Westland describes the use of bacterial cellulose in a fuel cell anode or a fuel cell cathode as specifically recited in claim 12. Thus, all of the limitations of claim 12 (and claims 13-16 and 21-23 that depend thereon) are not taught in any combination of Yoshitake, Kinsley, Westland and WO 89/12107.

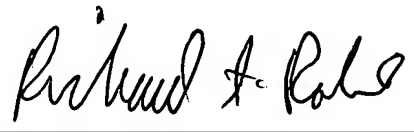
Conclusion

It is submitted that the entire application has been placed in condition for allowance. Favorable reconsideration is respectfully requested. A fee sheet is attached for the extra three claims. No other fees are believed to be needed for this amendment. If additional fees are needed, please charge them to Deposit Account 17-0055.

Respectfully submitted,

Barbara R. Evans *et al.*

Dated: February 6, 2004

By: 
Richard T. Roche
Registration No. 38,599
Quarles and Brady LLP
411 East Wisconsin Ave.
Milwaukee, WI 53202
(414) 277-5805

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ABSTRACT OF THE DISCLOSURE

The employment of metallized bacterial cellulose in the construction of fuel cells and other electronic devices is disclosed. The fuel cell includes an electrolyte membrane comprising a membrane support structure comprising bacterial cellulose, an anode disposed on one side of the electrolyte membrane, and a cathode disposed on an opposite side of the electrolyte membrane. At least one of the anode and the cathode comprises an electrode support structure comprising bacterial cellulose, and a catalyst disposed in or on the electrode support structure.

cellular plastic. A thermosetting or thermoplastic foam composed of cellular cores with integral skins having high strength and stiffness. The cells result from the action of a blowing agent, either at room temperature or during heat treatment of the plastic mixture. The resulting product may be either flexible or rigid, the latter being machinable. The foaming action in some cases may occur *in situ* (foamed-in-place plastics). Cellular plastics are combustible. For details, see foam, plastic.

Use: (Flexible) Furniture, automobile interiors, mattresses, etc., where softness and resiliency are desired. (Rigid) Insulating material, boat building and similar light construction, salvage of waterlogged ships.

See also foam, plastic; rubber sponge. For further information, refer to Cellular Plastics Division, Society of the Plastics Industry, 355 Lexington Ave., New York, NY 10017.

cellulase. An enzyme complex produced by the fungi *Aspergillus niger* and *Trichoderma viride* which is capable of decomposing cellulosic polysaccharides into smaller fragments, primarily glucose. It has been used as a digestive aid in medicine and in the brewing industry. Research has been devoted to experimental application of cellulase to disposal of cellulosic solid wastes. The resulting glucose can be fermented to ethanol, used to grow yeast for animal-feed proteins, or used as a chemical feedstock.

Note: Cellulase derived from the thermophilic soil fungus *Thielatia terrestris* functions at a much higher temperature than other types and is thus much more effective in decomposing cellulose. This indicates its possible use in conversion of biomass to energy. Commercial development of this product is expected.

See also biomass.

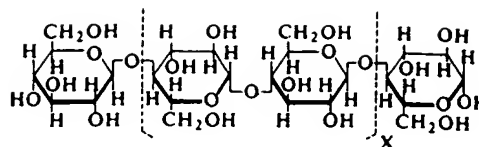
"Celluloid" [Hoechst Celanese]. CAS: 8050-88-2. TM for a plastic consisting essentially of a solid solution of cellulose nitrate and camphor or other plasticizer plus a flame retardant such as ammonium phosphate to minimize flammability; available in sheets, rods, tubes, films. Also called pyroxylin.

Hazard: Flammable, dangerous fire risk.

See also nitrocellulose.

cellulose. CAS: 9004-34-6. $(C_6H_{10}O_5)_n$.

A natural carbohydrate high polymer (polysaccharide) consisting of anhydroglucose units joined by an oxygen linkage to form long molecular chains that are essentially linear. It can be hydrolyzed to glucose. The degree of polymerization is from 1000 for wood pulp to 3500 for cotton fiber, giving a molecular weight from 160,000 to 560,000.



Cellulose is a colorless solid, d approximately 1.50, insoluble in water and organic solvents. It will swell in sodium hydroxide solution and is soluble in Schweitzer's reagent. It is the fundamental constituent of all vegetable tissues (wood, grass, cotton, etc) and is the most abundant organic material in the world. Cotton fibers are almost pure cellulose; wood contains approximately 50%.

The physical structure of cellulose is unusual in that it is not a single crystal, but consists of crystalline areas imbedded in amorphous areas. Chemical reagents penetrate the latter more easily than the former. Cellulose is virtually odorless and tasteless and is combustible, with an ignition point of approximately 450F. In some forms it is flammable. For example, railroad shipping regulations require a "flammable" label on such items as "burnt fiber," "burnt cotton," "wet waste paper," and "wet textiles." Fires have been known to occur in warehouses in which telephone books were stored. These were undoubtedly due to heat buildup in the paper caused by microbial activity and self-sustaining oxidation.

See also flammable material.

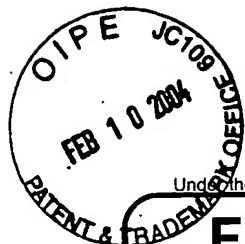
The most important uses of cellulose are bulk woods of many kinds; paper, most of which is made from wood pulp; cotton products (clothing, sheeting, industrial fabrics); packaging, ranging from wooden barrels to candy pats; and as a source of ethanol (enzymatic hydrolysis) and methanol (destructive distillation of wood). Specialized uses include nonwoven fabrics, medical equipment (artificial kidney), insulation and soundproofing, sausage casings, etc. Cellulose has approximately 60% of the energy content of bituminous coal; its use as a fuel has increased, especially in rural locations.

See also biomass.

There are many chemical modifications of cellulose, including its esters (cellulose acetate), ethers (methylcellulose), the nitrated product (nitrocellulose), and rayon and cellophane (from cellulose xanthate). Thus, it is the basis of many plastics, fibers, coatings, lacquers, explosives, and emulsion stabilizers. Alkali cellulose is an intermediate made by the action of sodium hydroxide solution on cellulose and is used for making cellulose ethers and viscose.

See also cellulose, modified.

Cellulose exists in three forms— α , β , and γ . α -cellulose has the highest degree of polymerization (DP), and is the chief constituent of paper pulp. It is insoluble in strong sodium hydroxide



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FEE TRANSMITTAL for FY 2004

Effective 10/01/2003. Patent fees are subject to annual revision.

☐ Applicant claims small entity status. See 37 CFR 1.27

TOTAL AMOUNT OF PAYMENT (\$) 54.00

Complete if Known

Application Number	10/017.202
Filing Date	12-14-2001
First Named Inventor	Barbara R. Evans
Examiner Name	Raymond Alejandro
Art Unit	1745
Attorney Docket No.	920976.90199

METHOD OF PAYMENT (check all that apply)

☐ Check ☐ Credit card ☐ Money Order ☐ Other ☐ None

☒ Deposit Account:

Deposit Account Number: 17-0055
Deposit Account Name: Quarles & Brady LLP

The Director is authorized to: (check all that apply)

☒ Charge fee(s) indicated below ☒ Credit any overpayments

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☐ Charge fee(s) indicated below, except for the filing fee to the above-identified deposit account.

FEE CALCULATION

1. BASIC FILING FEE

Large Entity	Small Entity	Fee Description	Fee Paid
Fee Code (\$)	Fee Code (\$)		
1001 770	2001 385	Utility filing fee	
1002 340	2002 170	Design filing fee	
1003 530	2003 265	Plant filing fee	
1004 770	2004 385	Reissue filing fee	
1005 160	2005 80	Provisional filing fee	
SUBTOTAL (1)			(\$) 0.00

2. EXTRA CLAIM FEES FOR UTILITY AND REISSUE

Total Claims	Extra Claims	Fee from below	Fee Paid
23	-20* = 3	x 18	= 54.00
Independent Claims	5	-5** = 0	x = 0.00
Multiple Dependent			

Large Entity	Small Entity	Fee Description
Fee Code (\$)	Fee Code (\$)	
1202 18	2202 9	Claims in excess of 20
1201 86	2201 43	Independent claims in excess of 3
1203 290	2203 145	Multiple dependent claim, if not paid
1204 86	2204 43	** Reissue independent claims over original patent
1205 18	2205 9	** Reissue claims in excess of 20 and over original patent

SUBTOTAL (2) (\$) 54.00

**or number previously paid, if greater; For Reissues, see above

FEE CALCULATION (continued)

3. ADDITIONAL FEES

Large Entity	Small Entity	Fee Description	Fee Paid
Fee Code (\$)	Fee Code (\$)		
1051 130	2051 65	Surcharge - late filing fee or oath	
1052 50	2052 25	Surcharge - late provisional filing fee or cover sheet	
1053 130	1053 130	Non-English specification	
1812 2,520	1812 2,520	For filing a request for <i>ex parte</i> reexamination	
1804 920*	1804 920*	Requesting publication of SIR prior to Examiner action	
1805 1,840*	1805 1,840*	Requesting publication of SIR after Examiner action	
1251 110	2251 55	Extension for reply within first month	
1252 420	2252 210	Extension for reply within second month	
1253 950	2253 475	Extension for reply within third month	
1254 1,480	2254 740	Extension for reply within fourth month	
1255 2,010	2255 1,005	Extension for reply within fifth month	
1401 330	2401 165	Notice of Appeal	
1402 330	2402 165	Filing a brief in support of an appeal	
1403 290	2403 145	Request for oral hearing	
1451 1,510	1451 1,510	Petition to institute a public use proceeding	
1452 110	2452 55	Petition to revive - unavoidable	
1453 1,330	2453 665	Petition to revive - unintentional	
1501 1,330	2501 665	Utility issue fee (or reissue)	
1502 480	2502 240	Design issue fee	
1503 640	2503 320	Plant issue fee	
1460 130	1460 130	Petitions to the Commissioner	
1807 50	1807 50	Processing fee under 37 CFR 1.17(q)	
1806 180	1806 180	Submission of Information Disclosure Stmt	
8021 40	8021 40	Recording each patent assignment per property (times number of properties)	
1809 770	2809 385	Filing a submission after final rejection (37 CFR 1.129(a))	
1810 770	2810 385	For each additional invention to be examined (37 CFR 1.129(b))	
1801 770	2801 385	Request for Continued Examination (RCE)	
1802 900	1802 900	Request for expedited examination of a design application	

Other fee (specify) _____

*Reduced by Basic Filing Fee Paid

SUBTOTAL (3) (\$) 0.00

SUBMITTED BY

Name (Print/Type)	Richard T. Roche	Registration No. (Attorney/Agent)	38,599	Telephone	414-277-5805
Signature	<i>Richard T. Roche</i>	Date	2-6-2004		

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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁴ : C12P 19/04, C12R 1/01, 1/02 C12R 1/05, 1/38, 1/41	A1	(11) International Publication Number: WO 89/12107 (43) International Publication Date: 14 December 1989 (14.12.89)
(21) International Application Number: PCT/US89/02355 (22) International Filing Date: 30 May 1989 (30.05.89) (30) Priority data: 199,606 31 May 1988 (31.05.88) US (71)(72) Applicant and Inventor: BROWN, R., Malcolm [US/ US]; 305 Skyline Drive, Austin, TX 78746 (US). (74) Agent: PAUL, Thomas, D.; Fulbright & Jaworski, 1301 McKinney, Houston, TX 77010 (US). (81) Designated States: AT (European patent), AU, BE (Euro- pean patent), BR, CH (European patent), DE (European patent), DK, FI, FR (European patent), GB (European patent), IT (European patent), JP, KR, LU (European patent), NL (European patent), NO, SE (European pa- tent).		Published <i>With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>
(54) Title: MICROBIAL CELLULOSE AS A BUILDING BLOCK RESOURCE FOR SPECIALTY PRODUCTS AND PRO- CESSES THEREFOR (57) Abstract The production of articles from bacterial cellulose is disclosed. A novel process also is disclosed for manufacturing bacteri- al cellulose which, in turn, is useful for producing a variety of articles.		

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MICROBIAL CELLULOSE AS A BUILDING BLOCK
RESOURCE FOR SPECIALITY PRODUCTS AND
PROCESSES THEREFOR

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BACKGROUND OF THE INVENTION

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This invention relates to a method of manufacturing for a multitude of new speciality products utilizing microbial cellulose. The present invention also relates to the products made according to this method. Ultrathin-superstrong transparent films and tissue growth medium articles/compositions, especially self-supported and with nutrients incorporated therein, are outstanding examples of products according to the present invention. Speciality papers, transport media, chemical derivatives, etc., from microbial gels are also features of the invention described herein.

35

The prior art has generally recognized that gels formed of microbial-produced-cellulose microfibrils (pellicles) are essentially usable directly for certain end-use and product applications in which the cellulosic characteristic of microbial-produced cellulose

1 microfibrils is applied as a substitute for conventional
cellulose. Copending United States Application No. 684,
844, filed December 21, 1984, is an illustration of an in
situ utilization of the cellulose forming ability of
5 certain microbes.

Examples of this substitution made in the prior
art include U. S. Patent 4,588,400 in which microbial
produced cellulose microfibrils (MC) pads are used to
retain medical fluids analogously to a cotton pad or a
10 fabric holding liquids. Also, the present inventor's
United States Patent No. 4,378,431 utilizes the cellulosic
character of microbial-produced cellulose microfibrils to
coat other fibers and fabrics to impart a bulk cellulosic
characteristic to the surface thereof. Thereby, articles
15 composed of such coated fibers have the feel, dyeability,
printability, liquid sorbtion and other characteristics of
cotton fabrics. European Application NO. 0228779 is an
example of a process patent directed to a reticulated
fibril configuration.

20 While the above-noted patents reflect quite
recent developments in this area, further advances have
been made. These advances have resulted in improved
cellulose product and ultimately improved or completely
new applications of same in end products.

25 SUMMARY OF THE INVENTION

The generic essence of the present invention is
the discovery that improved microbial-produced, cellulose
30 microfibrils (MC) can be produced and used as a much more
versatile intermediate and starting material building
block than previously realized, in order to form numerous
new classes of unique derivative products. These products
have properties and characteristics not obtainable from
35 conventional large fiber cellulose and are not
contemplated by the prior MC art.

1 The process methodology for relating microbial
cellulose properties with end-use products is also a
feature of this invention.

5 Thus, the broad scope of the instant invention
comprehends the processes for making such novel products,
the products per se, as well as the processes for using
them. In many instances the use for a particular product
describes the utility for the unique processes and
products of the invention, all of this being detailed more
10 fully below as best known inventive embodiments.

DESCRIPTION OF PREFERRED EMBODIMENTS

15 Although many specific embodiments will be
described herein, it will be apparent that part of the
methodology of the invention is the discovery of improved
methods of producing microbial cellulose from
micro-organisms. The resulting cellulose has a large
number or set of properties and processes which can be
20 collected and compiled for transposing microbial-produced
cellulose microfibrils into compositions, articles, and
structures having nonobvious properties, not previously
known for cellulosic compositions.

25 Microbial cellulose, as a substitute for
conventional cellulose and for applications in which
conventional cellulose was not previously used, finds a
variety of uses. The following list is exemplary:

30 (A) nonwovens and films, including speciality
papers, filtration and separation media, also including
membranes that could be used for water purification, films
on which inorganic films can be deposited and the like;

 (B) a speciality carrier, such as for battery
fluid and fuel cells;

35 (C) materials having special electronic effects
produced by coating the individual microbial-produced,

1 cellulose, microfibrils with appropriate components, such
as metals by vapor deposition or epitaxial growth;

(D) carriers for body-related materials, such as
5 foods, cosmetics, skin/hair treating materials and
internal drugs;

(E) mixing agents and viscosity modifiers per se
or as chemically and physically modified, in applications
such as surface coatings, particularly paints and fillers,
plasters, glues, adhesives, grouts and caulks;

10 (F) new specialty fibers, such as carbonized
versions, which can be used as polymer fillers and also
noncarbonized fibers, especially when strength and
biodegradability are desired;

(G) light transmitting optical fibers;

15 (H) wavelength and other electromagnetic and
radiation modifying materials;

(I) microfiber blends, especially with
melt-blown and other polyolefin fibers and in admixture
with many other different types of fiber to achieve
20 special effects and biodegradability;

(J) substrate media, especially self-supported
forms for growing plant and animal tissue;

(K) foods, food substrates and good fiber
substitutes;

25 (L) speciality laboratory uses such as for
testing for cellulase activity and substrates for
biological separations;

(M) other shapes such as specialty clothing,
which is lint-free;

30 (N) special property-modifying process steps and
modifications;

(O) synthetic leather and other texturized and
special appearance surfaces;

35 (P) diet fiber substitutes, such as for psyllium
fibers and in admixture with other dietary fibers, such as

1 wheat and oat (MC has attributes of soluble fiber because
of its submicron dimensions);

(Q) blends with other fibers, such as cotton as
a substitute for synthetics, such as polyesters and
5 nylons, in woven fabrics and nonwoven articles; and

(R) moisture-absorbing soil-enhancing additives
and conditioners.

The processes by which microbes produce
microbial-produced-cellulose-microfibrils are well-known
10 to the art. In general, the technique is described in the
inventor's previously issued U. S. Patent 4,378,431, the
disclosure of which is hereby incorporated by reference.

Any microbial strain capable of generating
cellulose is generally usable for the processes, articles
15 and compositions of the invention. These will be
generically referred to as cellulose producing microbes
(M). More specifically, those in the Acetobacter,
Rhizobium, Agrobacterium and Pseudomonas genera, as
described by the present inventor in his article in the J.
Applied Science: Appl. Polymer Symp. (1983) 37, 33-78,
20 the disclosure of which is hereby incorporated by
reference, are preferred. The species Acetobacter xylinum
is particularly preferred.

A novel discovery and part of the unique process
25 covered by the present application is that improved
cellulose results from the particular selection of microbe
strains which are capable of reversing direction of travel
during cellulose synthesis. Such a reversal results in a
more dense and stronger cellulose. Particularly preferred
30 is the NQ-5 strain of Acetobacter xylinum (American Type
Culture Collection 53582). A more detailed explanation of
this phenomenon is described in co-pending U.S.
Application No. 023,336, filed March 9, 1986, and
entitled, "Multi-Ribbon Microbial Cellulose", the
35 disclosure of which is hereby incorporated by reference.

1 Growth of the cellulose is promoted by a culture
medium which contains the microbes. The major constituent
of the culture medium for Acetobacter is a soluble
saccharide, particularly sugars, most particularly a
5 hexose and especially glucose.

Suitable nutrients are well known to the art.
One known as Schramm & Hestrin medium is especially
preferred. It generally comprises about 20 g/l glucose, 5
g/l peptone, 5 g/l yeast extract, 2.7 g/l anhydrous
10 diabolic sodium phosphate, and 1.15 g/l citric acid
monohydrate. Corn steep liquor and molasses are practical
and inexpensive sources of the hexose component preferred
in the nutrients of the invention. Another satisfactory
nutrient composition comprises about 8 volume percent
15 vinegar, 5 volume percent ethanol and 4 weight percent
malt extract. The pH is preferably adjusted to about 3 to
6, most preferably about 3.5 to 5.5. When it is desired
to increase the amount of oxygen-containing components in
the nutrient, additional alcohols and mixtures thereof can
20 be included in the nutrient.

The ambient temperature for maximum effectiveness
of microbial cellulose production is about 15 to 40,
preferably about 20 to 30 degrees Centigrade. The total
amount of time needed for acceptable cellulose production
25 is generally from about 1 to 25 days. Techniques for
improving microbe growth and increased cellulose
production from each microbe are contemplated by this
invention.

In copending, United States Application No.
30 684,844, filed December 21, 1984, and entitled "Production
of Microbial Cellulose", the disclosure of which is
incorporated herein by reference, on which the instant
inventor is a coinventor, a comprehensive inventive scheme
is disclosed for utilizing cellulose producing microbes
35 for producing shaped cellulosic objects on or within a

1 template. Various chemical and physical modifications are
disclosed to enhance and improve such shaped objects.
Similarly, this inventor's U. S. Patent No. 4,378,431
utilizes an existing fibrous structure as a
5 template/substrate for depositing a layer of cellulose in
situ from cellulose producing microbes. This approach
essentially utilizes the shape-forming ability of
cellulose producing microbes to form shapes that could
also have been formed from slurries of cotton/conventional
10 cellulosic fibers, even though not so practical a process
as that based on cellulose-producing microbe techniques.

The conventional product of cellulose-producing
microbes is a mass of intertwined ribbons comprised of
cellulosic microfibrils. These ribbons are generated at
15 the oxygen-containing gas (air is operable)-nutrient
interface. This mass is translucent, insoluble, but very
hydrophilic and wettable and has great tensile strength.
It appears to be a gel to sight and touch. It, as well as
products made therefrom, have exceptionally high dry
20 tensile strengths and dimensional stability.

One of the features of this invention is that the
oxygen/liquid nutrient interface can be conveniently
obtain by growing the microbes in an enclosed plastic film
or bag. Each such container is a discrete reactor and can
25 be designed to be of any size not exceeding the bursting
strength of the container or its sealing means. It is
preferably oblong, about 0.5 to 2, preferably about 0.75
to 1.5 feet in width and about 0.25 to 0.6, preferably
about 0.3 to 0.5 feet in height. It should be at least
30 about 0.5 feet in length.

Air space is provided above the liquid surface in
the container. It is desirable to utilize a plastic film
having the characteristic of a relatively high oxygen
diffusion ability with low permeability to the liquid
35 nutrient molecules. This practical, versatile reactor can

1 be designed for any location and has particular
applicability in remote areas. If desired, the
oxygen-containing environment within the reactor can be
increased to improve oxygen availability to the microbes
5 and, thereby, obtain a more efficient cellulose conversion.

In addition to the foregoing advantages, these
reactors are desirable because the contamination problems
ordinarily plaguing biotech processes can be easily
controlled and eliminated. While these reactors have been
10 described in connection with cellulose producing microbial
processes, they are intended to be used in any bioprocess
in which their usefulness can be enjoyed.

Another feature of the invention is the
recognition that an improved cellulosic product is
15 obtained by adding an agent to the nutrient bath which
interferes with crystallization, but not polymerization,
of the cellulose. Suggested agents include dextran having
substituent groups such as alkyl, alkyl carboxyl,
alkylhydroxyl, sulfate, sulfonic acid or alkylphosphate.
20 Particularly preferred is carboxymethyl-cellulose (CMC).
This concept is described in more detail in United States
Application No. 022,904 filed March 6, 1987, and entitled
"Microbial Cellulose Modified During Synthesis", the
disclosure of which is hereby incorporated by reference.

25 The generic concept of the present invention
transposes the improved microbial cellulose into novel and
nonobvious products, which utilize the special properties
of microbial cellulose produced according to the present
processes, which are not obtainable from other cellulosic
30 sources or other microbial cellulose produced to date.
One of the breakthrough, inventive concepts of the present
invention is the realization that the unique properties of
cellulose producing microbes can be collected, catalogued
and innovatively harnessed to customize unique products.
35 Certain final product properties are defined and those

1 corresponding properties are selected so that microbial
cellulose is adopted and tailored to be utilized in a huge
variety of processes, products and compositions having no
counterpart in the prior art.

5 The cellulose microfibrils produced from microbes
have submicron cross-sectional diameter dimensions of from
1.5nm (nanometers) [0.0015 micron] to 10nm (0.01 micron)
This results in an enormous fiber surface area per cubic
volume of fiber. Moreover, the submicron dimensioned
10 cellulose fibrils produced by microbes having
exceptionally high wet and dry tensile strengths. These
microfibrils are especially noteworthy with respect to
their remarkably high length to diameter ratio which can
be in the order of as much as millions to one.

15 Dispersions of wet submicron fibers can be wet
spun or pulled into larger fibers or yarns of filaments of
exceptional high tensile strength and Young's Modulus.
This can be accomplished with the pure MC cellulose
fibrils, as well as chemical, genetic and physical
20 modifications thereof, both before and after the process
of producing the larger materials from the submicron
fibrils.

In addition, the fibers, filaments or yarns can
be carbonized. They have exceptional strength because
25 parallel molecular orientations can be obtained.
Polyacrylonitrile (PAN) fibers are currently the choice of
the art for maximum strength. Unmodified microbial
cellulose can be carbonized to approximate these
properties. Further, grafts of acrylonitrile can be made
30 to microbial cellulose. That grafted product will be a
composite of the preferred properties of microbial
cellulose and PAN for a preferred starting material for
fiber carbonization.

35 These PAN grafts have inordinately high water
absorption capabilities. The Department of Agriculture

1 recently patented a PAN-starch graft or copolymer which
purportedly will absorb up to 1,000 times its weight of
water. Microbial cellulose-PAN (MC-PAN) has at least
comparable absorption properties; and, because of its
5 fibrous characteristic, it can be used in environments in
which structural integrity, as well as wettability, is
important. For instance, an irrigating hose formulated
from MC-PAN would constantly drip water from a saturated
state. The tissue growing aspects described later herein
10 will be enhanced in some aspects by the used of MC-PAN.

Although a dried pellicle or film from cellulose
producing microbes, has some paper-like aspects, the
invention goes beyond that primitive level and advances
the state of the paper/non-woven and film art
15 dramatically. The key is that microbial cellulose
according to the present invention has greatly different
physical characteristics than conventional cellulosic
fibers. This factor is inventively utilized to select
certain types of microbial cellulose-based articles, such
20 as specialty papers, that especially benefit from those
special microbial cellulose properties. One of these is
speciality paper, particularly those that need to be free
of inorganic acids to avoid degradation.

Such papers include formal documents exemplified
25 by diplomas, treaties, certificates and the like. These
will be superior in aging, bending-resistance, tear
resistance and other strength factors. Some of the
process embodiments described elsewhere in this
application, such as glycerol and CMC, can be used to
30 enhance these properties. This utility is described in
more detail in United States Application No. 199,780,
filed May 31, 1988, and entitled "Microbial Cellulose
Composites and Structures from in situ Formation".

Thus, in those paper, film membrane and other
35 related applications, where flexibility and bending

1 strength, particularly at low temperatures, are desired,
the MC can be treated with glycerol to obtain a vast
improvement in these already respectable capabilities of
MC.

5 In particular, the assemblage of submicron range
diameter microfibrils results in a paper that also has an
outstanding ability to accept inks, dyes, toner and other
color impressions resulting in images of far greater
resolution than is possible with conventional cotton
10 linters, rag or wood based papers.

Moreover, the high quality paper as described
above is specially adapted to be coated with photographic
emulsions to produce photographs of very fine grain and
definition. This permits the elimination of resin coating
15 which is ordinarily utilized to mask the roughness of
conventional papers.

To much the same effect, the microbial cellulose
substrate can be coated with magnetic media and media
capable of deformation often by heat for optical reading
20 purposes under laser light. In addition to the
receptivity and adherability of magnetic and photographic
coatings, the strength and dimensional stability of
microbial cellulose, especially at temperature extremes,
all contribute to a superior support material.

25 The multiplicity of submicrobial fibers in a
dry-state article can be coated with appropriate
substances, especially in thicknesses as thin as single
molecular layers of various materials, such as,
conductors, to make the entire article electrically
conducting. In some instances the selection of deposition
30 material is effected so as to achieve any of
superconductive, ordinarily conductive or semiconductor
properties. Furthermore, the dry ribbons and thin films
of MC in either the coated or noncoated stage can be
oriented and adjusted to achieve different absorption
35 properties for light and electromagnetic radiation.

1 Microbial cellulose paper and three dimensional
dry industrial applications articles are especially
suitable for certain industrial applications. These uses
include:

- 5 (a) as a base for a polymerizable monomer or
resin impregnated for such uses as circuit
boards, friction discs for transmission
plates and any application where strength
and superior impregnability factors are
10 important. MC in dispersed fiber form can
be used as a reinforcing agent in a wide
variety of composite articles; and
(b) diaphragms to be vibrated for sound
transmission in both set and dry
15 environments, such as those in loudspeakers,
earphones, telephone transmitters. These
diaphragms may be metal-coated for enhanced
properties.

Resin impregnated or coated MC films, paper, or
20 other shape can be built up into three dimensional
thermosetting resin articles, where they will have great
extreme temperature stability and useability. One
excellent unique application is to form the molded nose
cone of rockets, where they will provide ablative barriers
25 for heat-resistance to atmospheric friction. Also in
submarine nose cones, they will provide the resistance to
pressure and temperature extremes and be used to house
sonar generating equipment without metal barriers.

Even more unusual is the ability of diaphragms,
30 membranes and films of microbial cellulose to be used
under water, not only for sonar devices, but for
underwater sound applications yet to be developed because
no membrane of this type has been hitherto available. In
hot water, the dimensional stability of these materials is
35 exceptionally useful.

1 The ability of microbial cellulose structures,
such as membranes and films to retain their structural
integrity while wet and under water is especially
important in many membrane applications. For instance, in
5 the human body, kidney and heart implant membranes will
play useful roles, especially since extremely thin and
small articles can be used because MC has such strength
and stability even in ultrathin cross sectional forms.
And outside the body, blood and other body fluids can be
10 treated and filtered. Water purification, such as by
reverse osmosis on both small scale personal as well as
large scale industrial situations, is well-suited for MC
membranes, etc. Other separations where small pores from
submicron microbial fibers accompanied by unusual strength
15 are important factors can be achieved by microbial
cellulose in appropriate shapes and configurations.

 Moreover, suitably shaped three-dimensional MC
items can be used as surgical implants in both gel and dry
forms. In the dry form, it provides a multiplicity of
20 interstices in which body tissue can renew itself for good
healing and bonding. It is likely that MC will not
stimulate extreme body rejection mechanisms, but it can be
impregnated with cyclosporin and the like to minimize
rejection problems.

25 Also, the microbial cellulose paper can be
employed in a transparent film, which is molded or cast to
be used as negatives and color transparencies in the
graphics and reproduction industries.

 In addition to the high strength and modulus
30 generally characterizing the products of this invention,
special mention must be made of the exceptional
dimensional stability of these products, both at extreme
high and low temperatures and with respect to very thin
planar film or membrane forms, as well as small
35 cross-sectional three-dimensional shapes.

1 One exceptionally unusual discovery is that
remarkably thin films can be cast from dispersions of
microbial cellulose, particularly when treated with
glycerol or CMC or both. These films have a thickness of
5 less than about 0.1 micron, e.g. 80 nanometers or 0.08
microns. The unusually thin films have remarkable
strength and dimensional stability. Before drying they
can be stretched for orientation to further enhance
strength and stability properties. Moreover, they and
10 other film/membranes of the invention can be twisted into
filaments/threads/fibers, also with outstanding
strength/stability properties.

 It has been further discovered that inorganic
materials, such as metals can be vapor deposited or
15 epitaxially grown in approximately monomolecular layers on
the surface of these ultra thin planar/filamentous
materials. Thus, conducting, semiconducting and
superconducting materials can be formed in such ultra thin
layers on these ultra thin microbial cellulosic structures.

20 Since microbial cellulose is capable of remaining
flexible and stable at temperatures as low as those of
liquid helium, laminar composites of microbial cellulose
and films of superconductive materials offer considerable
promise in providing flexible superconductor conduits and
25 structures that are not possible to achieve with the
brittle, almost-ceramic, new superconductors that have
recently excited the scientific/industrial community.

 The May 24, 1988 Wall Straat Journal reported
that thin layers of the new thallium superconductor are
30 adequate conductors of electricity for some commercial
applications. It was demonstrated to conduct 110,000 amps
per square centimeter. The layers are presently being
deposited on silicon. Depositing on microbial cellulose
substrates according to the instant invention, opens the
35 door to an enormous potential for flexible electronic

1 components at extremely low temperatures and/or in
exceptionally small housings.

Films of this microbial cellulose are also
excellent for use as edible casings, such as those used
5 for sausages and hot-dogs.

The properties delineated above also lend
themselves to microbial cellulose papers that serve as
excellent wall coverings. They can be easily texturized
and provide the strength of vinyl wall coverings without
10 any loss of breathability of the wall, thereby obviating
unwanted vapor barriers in a room.

These properties of a microbial cellulose are
also applicable to electrical insulating applications,
especially when very high electrical power loads are
15 involved. They are also equally applicable for thermal
insulating environments. Any shape can be fabricated.
Non-melting and cold-insensitive microbial cellulose
structures are particularly suitable in extreme
temperature environments.

20 These same insulating and breathing properties
are vital to achieve special benefits for clothing to be
used in specially harsh environments, such as the space
program. In that connection, the extra-vehicular activity
suit, as well as the ones worn on the space vehicle must
25 be lint-free. That is a special property of microbial
cellulose.

This type of clothing, including hand-gloves
where suitable, must be pressure and puncture resistant,
again a special property of microbial cellulose.
30 Moreover, the gloves and clothing retain their flexibility
at extremely low temperatures. They are also useful for
civilian applications such as skiing and mountain climbing
clothing.

All clothing items and insulators can be formed
35 by in situ means, such as described in U.S. Application
No. 684,844.

1 Special texturized and insulating effects can be
obtained by freeze-drying a pellicle. The surface of the
resulting product appears leather like. A formed layer
below the surface contributes to the excellent insulating
5 properties and uses of this material. This would apply
not only to clothing but also for such items as shoes and
boots. This texturing can also be used as one of the
tools for achieving visual effects in the artificial food
embodiment of this invention.

10 In U.S. Patent No. 4,588,400, the disclosure of
which is hereby incorporated by reference, a microbial
cellulose pellicle pad loaded with physiologically-
acceptable liquids for medical applications is disclosed.
It is part of the instant invention to advance and expand
15 the limited scope of this patent to include cosmetics,
soaps, skin-cleaning and hair treating agents. This
expansion would also include drugs to be absorbed directly
by the skin.

Also, not contemplated by the '400 patent is
20 encapsulating the drug with MC according to a preselected
configuration to deliver drugs to a distant part of the
digestive system without the drug first having been unduly
and prematurely exposed to the digestive liquids of the
mouth or stomach.

25 Microbial cellulose has special usefulness as
gums and gels (such as xunthan or algerate), which are
well-known classes of materials used for a wide variety of
applications. Microbial cellulose pellicles and other
variants can be used in any of these known applications.
30 In most instances they will impart superior properties in
these environments. One reason is because of their
self-supporting properties and characteristics.

Notwithstanding the well-established gum/gel art,
microbial cellulose has been found to far exceed the
35 capabilities of agar as a tissue growth medium. This is

1 because the huge dispersion of submicron hydrophylic
cellulose fibers constituting the typical microbial
cellulose pellicle is a most superior tissue growing
medium for both animal and plant tissues. There are
5 several reasons for this outstanding capability. One is
that microbial cellulose gels have exceptional structural
strength. Two, they provide a perpetually wet medium for
hair roots without drowning them and at the same time
ensuring the hair roots have an adequate oxygen supply.

10 These same characteristics also enable microbial
cellulose gels to perform outstandingly as seed coatings.
They can enhance and control germination and promote seed
development. Moreover, the seeds can be properly spaced
in predetermined gel configurations. The gel can be
15 packaged in a container to be dispersed by the user, so
that each individual seed can be encapsulated in gel at
the user's choice.

Plant tissue can be incorporated into the
microbial cellulose gel to obtain effective asexual
20 reproduction from various sources of growable tissues.

The microbial cellulose with seed or tissue or
plant suitably incorporated therein can be encased in
plastic film containers or tents to obtain a controlled
environment. This encompasses moisture control as well as
25 resistance to contaminants, such as viruses and other
pathogens. The microbial cellulose can be impregnated
with liquid fertilizer, fungicides, and insecticides to
further control the environment. The evaporation rate
will be particularly controllable, thereby enhancing the
30 longevity of the growing material. Furthermore, if the
plastic of the tent is selected to be relatively oxygen
permeable, even greater benefits are obtained.

The same type of approach will make these gels
useful in fuel cell and battery structure articles, where
35 the electrolyte comprises the liquid phase of the
microbial cellulose gel.

1 Further, a cellulose membrane (pellicle) can be
loaded with various cosmetics, skin treating compounds,
wrinkle removing compounds and other drugs, emollients,
5 hair treatments and hormones for the skin. The cellulose
membrane containing these ingredients is then placed on
the skin. The result is that a thin skin of microbial
cellulose forms as an outer layer and prevents evaporation
of the ingredient off the skin. This results in a longer
10 and more concentrated exposure of the ingredient on the
skin. Similarly, sun screen compositions and mud packs
can be enhanced. MC emulsions or suspensions made by
physically masticating MC in a mechanical shearing device,
such as a blender, can be used per se as a mud or blended
with conventional face mud formulations to enhance them.
15 These emulsions and suspensions have a wide variety of
other uses as will be apparent from the entirety of this
disclosure.

Only one established use of microbial cellulose
in pellicle form as a food is known. That is a simple
20 sugar flavored pellicle. The instant invention proceeds
considerably beyond that primitive state of the art.
Accordingly the present invention incorporates mouth-feel,
texture, shape, density and flavor parameters into
designing the microbial cellulose pellicle, or dispersion
25 or other gel or physical embodiment to achieve desired
effects and result in a particular designed food item.

As one example of this methodology, microbial
cellulose, can be obtained and molded or shaped into raw
oyster or clam shape/feel pellicles. Clam/oyster flavor
30 can be incorporated into pellicles of requisite
texture/shape to obtain very close approximations to the
natural material. Under the same focus, steaks and other
selected fish, fowl and animal food types can be
duplicated and artificial sausages, bacon can be
35 formulated. An imitation beef jerky can also be made.

1 Since cellulose is not digested, microbial
produced cellulose provides an ideal non-fattening bulking
medium that can be engineered into a wide variety of
physical forms and appearances. Accordingly, MC can be a
5 carrier for a wide variety of specialty selected flavors
and nutrients for the human or animal body. For instance,
it can carry flavors, dyes, fats, lipids, etc. In fact it
can be configured to taste and feel like a fat. Puddings,
ice creams, salad dressings, creams, spun confections can
10 all be formulated from microcellulose gels and
dispersions. Imitation vegetable oils, dressings,
mayonnaises, butters, sour creams, cottage cheeses, hard
and soft cheeses and spreads are other food variations of
viscous food embodiments of this invention.

15 The types of synthetic foods include:

1) Hard form--this includes artificial
potato chips, corn chips, nuts, such as peanuts, macadamia
nuts, walnuts, Brazil nuts, and other snacks. Hardness
can be accentuated by cross linking and/or prolonging the
20 production of a microbial cellulose to achieve very high
microfibril production and density.

2) Soft form--this includes artificial
mashed potato, noodles, spaghetti, rice granules,
tortillas and other Mexican food products, cake fillings,
25 fudge, candy bar fillings and the like.

3) Fillers--bakery products, artificial
flour, cereals, expanded encapsulated microbial cellulose
for puffed cereals and artificial popcorn.

4) Ice Nucleation Agents--added and
30 included in various proportions with all frozen foods to
control ice formation by nucleation or otherwise to
achieve special effects, such as creaminess in popsicles,
frozen fruit bars, ice creams, sherbets, and other frozen
foods and desserts. Because microbial cellulose
35 interferes with crystalline ice structure, it by itself or

1 in synergistic admixture with other ice crystal modifiers
is particularly useful as a component of liquids to be
frozen into smooth texture foods.

5 Of course, it will be desirable that some of the
foods to be formulated will not only be resistant to
crystalline ice formation, but also to melting (fudge,
butter.) Resistance to both such extremes is a
characteristic of microbial cellulose and can be designed
and engineered into the products of this invention.

10 These artificial foods can be made free of
deleterious products that sometime plague natural foods,
such as virus, bacteria, etc.

15 The moist stage gel is also useful for testing
for the activity of cellulases organisms or substances,
especially for the presence of celluloses. In addition,
UDPB/cellulose synthase complex on a substrate, such as
microbial cellulose gel, could accept glucose or even
sucrose to be converted directly into cellulose in vitro.

20 It is disclosed in copending U.S. Application
No. 684,844, and it is apparent that an immense range of
chemical derivatives and modifications can be effectuated
on fabricated microbial cellulose structures. The
invention herein contemplates a considerable,
unobviousness stepout from that state of the art.

25 One concept of this invention is to regard the
finely divided in situ submicron-sized cellulose fibers as
most ideal starting raw materials for the complete panoply
of chemical reactions leading to a wide range of
industrial chemical materials. The reactions described in
30 detail in the McGraw Hill Cellulose article for cotton,
etc. are considerably improved by utilizing microbial
cellulose in a finely divided dispersed state. Thus, an
important feature of this invention is to utilize finely
divided microbial cellulose fibers obtained either by
35 mechanical dispersion or by agitation during growth of the

1 microbial cellulose so that chemical reactions can occur
therewith in situ. A wide variety of industrial chemicals
are made much more efficiently with this approach, since
expensive process steps needed to prepare ordinary
5 cellulosic materials for chemical reactions are eliminated.

MC gels, modified with certain grafted side
chains, such as PAN, have enormous water-absorbing
capacity. Both these and the unmodified versions can be
used in agriculture, drilling muds and as thixotropic
10 components of compositions, such as those used for
enhanced underground pumping for oil recovery.

Another subclass of the invention comprises
process modifications, immediately before and during the
microbial-produced cellulose microfibrils production stage
and immediately thereafter. These modifications are
15 primarily designed to impact the use properties of the
microbial-produced cellulose microfibrils, rather than to
improve the economics and efficiency of the process,
although in some instances both goals will be obtained.

20 It is known that ambient, atmospheric oxygen is
adequate to obtain reasonable yields of MC. Nevertheless,
the efficiency of the oxygen uptake by cellulose producing
microbes can be improved. One approach is to increase the
concentration of oxygen available to the microbes. This
25 can be accomplished by increasing the volume percent in
the ambient gas environment. The other is to dissolve
oxygen in the nutrient sodium by physical means such as
bubbling or agitation (can form discrete modules of MC) or
by chemical compounds such as peroxides. This includes
30 the use of chemical substances such as alcohols from which
the microbes can obtain their oxygen needs from the food
supply. The pressure of the ambient oxygen can also be
increased with hyperbaric techniques.

It is known from previous work that the
35 properties of microbial cellulose can be substantially

1 modified by the in situ treatment with
carboxymethylcellulose(CMC). The effect of CMC is thought
to be that of causing and maintaining splaying of
micro-fibril ribbon assemblages. In this invention it has
5 been discovered that other materials can substitute for
CMC. One such material is polyethylene glycol (PEG),
which, in addition to affecting splaying, also
beneficially effects strength, flexibility, water, water
absorption, optical clarity and the like.

10 Ribbon splaying can also be accomplished by
including certain enzymes active to cellulose, such as
cellulase, in the growth medium. Thereafter, once
satisfactory splaying has been accomplished, CMC, PEG,
etc. can be added as a post reaction step to maintain the
15 splayed state.

The pellicle can also be subject to modification
by post-formation viscosity or friction reducing modifying
agents. For example, drying a cellulose membrane in the
presence of glycerol results in a paper-like product of
20 greatly reduced brittleness and which is exceptionally
flexible.

It is also a feature of the invention that
different species of bacteria capable of producing a wide
variation of cellulose types will be systematically
25 selected to make the cellulose of choice by direct
synthesis. If existing strains of bacteria are unable to
produce a cellulose of a desired character, the strain can
be mutated by genetic or environmental techniques. Thus,
Acetobacter has been modified to produce cellulose acetate
30 directly. Other modifications leading to the biosynthesis
carboxymethylcellulose and other cellulose derivatives are
feasible.

35 Blends of MC with ordinary plastics will be
particularly important in applications requiring a high
degree of biodegradeability. An illustrative example is
provided U.S. Senate S. 1986 providing that plastic
six-pack yokes be degradable.

1 Capillary electrophoresis is highly promising for
rapid and accurate separations of ionic species, i.e.
amino acids, peptides, nucleic acids and the like. Many
capillary electrophoresis separations based on molecular
5 charge, such as isoelectric focusing, isotachophoresis and
micellar electrophoresis are useful. These rely on
gel-filled capillaries. The MC gels of this invention are
exceptionally well suited for this application, especially
since the gels can be prepared in situ in the capillary,
10 if that is appropriate for the separation need.

The filter/membrane/film form and separation
capabilities of the MC are especially useful in vapor
phase because of the extraordinary surface area of the
microfibrils. As an example, cigarette filters made of MC
15 are especially effective at removing adverse components of
tobacco smoke. MC can also be used on a much larger scale
as a filter to remove or control the level of tobacco
smoke in closed areas.

When pellicles or gels of MC are loaded with
20 nutrients, fertilizers, pesticides, fungicides, etc., they
can be very effective as long-term, slow release,
non-quick-drying units to be placed in areas of highest
effectiveness.

Gels, creams and pastes of a wide variety of
25 consistencies can be made and used from MC. All of these,
particularly when exposed to heat and cold, have a much
higher gel strength and integrity than standard for such
items. Industrial pastes and gels can be formulated as
rust removers, metal cleaners, foods, condiments, pastes,
30 and the like. Moreover, in addition to the
cosmetic-emolument and skin conditioners described above,
MC is an ideal hair conditioner and strengthener, because
the submicron fibrils can be associated with hair strongly
enough and in quantities high enough to have an
35 outstanding beneficial effect on hair appearance, body and
feel.

1 MC is a pure natural cosmetic. Many synthetic
conditioners contain chemical residues, such as benzene.
This makes them undesirable and in some instances unduly
toxic for cosmetic and other uses. Further MC is
5 exceptionally hypoallergenic (not allergenic).

Liposomes containing exceptionally small MC
fibrils as drug carriers can be injected into the body.
Ordinarily, liposomes are used to inject highly fatty
materials into the body that are often incompatible with
10 the body. The liposome compositions contemplated by this
invention can carry the submicron MC in the aqueous phase.

Synthetic sweeteners can be sorbed by MC which
can then be used as a carrier for a wide variety of frozen
and other foodstuffs.

15 A very high quality cellulose is sold
commercially as Microcrystalline Cellulose under the
trademark/trade name of AVICEL. This is made from a very
finely masticated high purity cellulose paper, such as
filter paper. The material is used as a component of
20 foods. It is not very soluble. Comparable physical forms
made from MC will be less expensive and superior in almost
every significant commercial property.

The present invention will now be further
illustrated by certain examples and references which are
25 provided for purposes of illustration only and are not
intended to limit the present invention.

Example 1

30 A Food Delicacy Made from Microbial Cellulose

MC (strain AY 201) was placed into standing
culture in a shallow 1 inch tray. The growth medium was
Schramm Hestrin Medium. The culture conditions were 28 C,
35 and after 2 weeks, a prominent gel-like membrane

1 (pellicle) appeared in and completely filled the culture
medium of the shallow culture tray. The pellicle was
removed, washed with distilled water, then cut into small
cubes about 1/2 inches square. The cubes were further
5 soaked in distilled water, then autoclaved at 240 C at
20 p.s.i. for 30 minutes, then rinsed with sterile
distilled water. Then the cubes were placed into an
aqueous solution saturated with ordinary table sugar
(sucrose). The cubes and sugar solution were then
10 autoclaved again at 240 C at 20 p.s.i. for 20 minutes,
cooled, then stored under sterile conditions until use.
The sugar cubes of MC were eaten as a delicacy. The mouth
feel and sweet taste imparted an excellent food delicacy.

15 Example 2

Sub-Micron Thin Cellulose Films

Strain NQ-5 of *Acetobacter xylinum* was
20 inoculated into Roux bottles containing 100 ml of Schramm
Hestrin Medium and cultured for 3 days at 28 C. At the
end of the second day, a thin, transparent pellicle formed
at the gas/liquid interface. This pellicle was harvested
and cleaned as follows: It was first soaked in distilled
25 water for 3 hours (3 changes), then in detergent (Alconox)
for 12 or more hours. Then the pellicle membrane was
thoroughly rinsed in distilled water to remove any
residual of detergent. The pellicle was then stretched of
the lip of a 250 mm glass beaker and allowed to air dry.
30 The resultant membrane was optically clear but exhibited
interference colors typical of thin films. The
interference colors suggested a dry pellicle membrane
thickness of approximately 100 nm. The pellicle membrane
is exceedingly strong and has great dimensional
35 stability. When heated to more than 100 C, there were no

1 apparent changes in the film. In one sample, electron
microscopy grids were placed on the pellicle membrane
before drying. Upon drying, the thin pellicle membrane
was stretched across the EM grids. The pellicle membrane
5 was then directly viewed in the transmission electron
microscope. These membranes consist only of one or two
layers of cellulose ribbons. The ribbon is the unit of
cellulose which emerges from the bacterium and is well
known in the literature. If stretching of the wet
10 membrane is isodiametric, the orientation of the ribbons
is random. If the wet membrane is stretched in two
preferable directions, ordered ribbons are produced.

Some of these sub-micron films were placed in a
15 high vacuum bell jar, and platinum was heated and
vaporized onto the surface. The resulting thin films were
electro-conductive and had great dimensional stability.

Example 3

20

Wet Spinning of Microbial Cellulose

A thin pellicle of microbial cellulose was
produced as described in Example 2. The cleaned pellicle
25 could be pulled by hand in two directions using a twisting
motion and also squeezing. These motions resulted in the
formation of a thin strong thread. Such threads could be
made as long as 6 inches. These threads can be used like
cotton fibers as starting material for yarn and textile
30 production. The advantages of MC threads is their
superior mechanical wet and dry strength as well as
continuous length of cellulose microfibrils. Threads as
small as 50 microns in diameter can be produced by the
above technique. During this pulling, the cellulose
35 ribbons co-align into parallel arrays, thus producing a

1 large number of intermolecular H-bonds between the
microfibril clusters. This property results in a superior
mechanical strength and dimensional stability of a fiber
for weaving.

5 In another thread making technique, silicon
tubing ranging in id diameter from 0.5 mm to 1.00 cm was
filled with Schramm and Hestrin Medium inoculated with
Acetobacter. After 3 days, microbial cellulose formed
10 within the tubes as exact casts of the silicon molds. The
cellulose was then extracted from the molds by pulling,
forming a strong cohesive thread. After pulling from the
mold, the thread was dried. This type of batch fed or
continuous biosyntheseis of threads will be a useful
15 fermentation method for producing high strength fibers for
the textile industry.

Example 4

20 Plant Seed Germination and Seedling Development in Microbial Cellulose

Microbial cellulose pellicles were produced in
trays as described in Example 1 above. The pellicles were
25 cleaned with 10% NaOH and detergent (Alconox), autoclaved
and rinsed with distilled water. Then the sterile
pellicles were inbibed with sterile inorganic nutrient
solution for plant seedling development. This solution,
known as Bold's Basal Medium, contains inorganic and
30 organic materials to grow photosynthetic organisms. Seeds
of radish (Sativis sp) were sterilized in household
bleach, rinsed in sterile distilled water, then placed on
the surface of the sterile pellicle which was soaked in
Bold's Basal Medium. Seedling germination was better than
35 90%, and the roots did not penetrate the microbial

1 cellulose substrate, but the root hairs grew into the
substrate. Seedling development by this technique is
superior to soil or other synthetic substrates since root
development into the substrate is hampered. Thus, the
5 young seedlings can be transplanted later into a more
disperse and open soil or synthetic substrate for
continued growth. Microbial cellulose is an excellent
medium to germinate delicate small seeds and spores from
ferns, mosses, fungi, etc. Thus it is anticipated to be
10 an excellent substrate for growing mushrooms.

Example 5

Synthetic Leather from Microbial Cellulose

15 A pellicle was grown as outlined in Example 1
above and cleaned with 10% NaOH and detergent, then
rinsed. The pellicle was then subjected to a standardized
freeze drying procedure whereby the water content, in the
20 form of ice, is sublimed from the pellicle. This leaves
the pellicle ribbons non-collapsed. The feel and strength
of this material resembles a fine patent leather good,
similar to that of fashionable hand gloves. Preservatives
can be added to the never dried material to prevent
25 enzymatic, chemical hydrolysis, or radiation damage to the
artificial leather.

Example 6

30 A Simplified Highly Efficient Fermentor for Microbial Cellulose

A tray similar to that described in Example 1
above was innoculated with Acetobacter. The entire
35 sterile tray was fitted into a previously sterilized

1 polyethylene bag which is permeable to oxygen. The bag
was inflated with air and sealed, then the culture was
allowed to synthesize cellulose. The cellulose yield with
strain NQ-5 exceeded 35%, and contamination was avoided.
5 The advantages of this system offer sterile environment
for bacterial growth and cellulose production in standing
culture. To improve the rate of cellulose synthesis, the
polyethylene bag itself has been filled with a thin layer
of liquid culture medium as described in Example 1, but
10 the bag was inflated with air or oxygen-rich atmosphere so
that the liquid surface was not in contact with the bag.
The advantages of this fermentor system is increased
culture surface area to the oxygen environment (the bottom
of the culture vessel was in direct contact with the
15 oxygen-permeable membrane) and the possibility for
increasing and controlling the oxygen content of the
atmosphere in association with the growing culture. This
technique can be modified for batch fed or continuous
fermentation of microbial cellulose.

20

Example 7

A Plasticizer Makes Dried Microbial Cellulose Membranes Less Brittle and Imparts Greater Strength

25

Microbial cellulose is produced and cleaned as in
Example 1 above, but it can be of any shape or form.
Before drying, the cleaned pellicle is soaked in a
distilled water solution containing 1-3% wt/vol glycerol.
30 After soaking for 24 hours while on a gyratory oscillator
to improve penetration, the pellicle is removed and then
air dried. The fully dried pellicle has different
physical properties from the pellicle dried only from
distilled water. The pellicle is very bendable and
35 resists tearing. It also has 50% greater Young's Modulus
in comparison with air dried clean cellulose.

Example 8

Resin Impregnated Microbial Cellulose

A never dried pellicle produced as described in Example 7 above was dehydrated over a 12 hour period in an ethanol/water series constituting of 25%, 50%, 75% and 100% ethanol. The ethanol soaked pellicle was then subjected to an acetone exchanges consisting of 25%, 50%, 75%, 100%, 100% over a 12 hour period. The pellicle was then infiltrated with a typical electron microscopy resin known as Spurr's resin. The infiltration series was 25% resin and 75% acetone for 3 hours; 50% resin and 50% acetone for 3 hours; 75% resin and 25% acetone for 12 hours; and, 100% resin for 12 hours, followed by a second exchange of 100% resin for 3 hours. The resin/pellice complex was heat polymerized at 65 C for 24 hours. The cellulose ribbon microfibrils imparts a greater strength to the resin. Flexibility is also increased.

Many modifications and variations of the present invention are possible in light of the above teachings. Therefore, it will be apparent to one of ordinary skill in the art that many changes and modifications can be made thereto without departing from the spirit or scope of the invention as set forth herein.

1 What Is Claimed Is:

1. An article of manufacture comprising
microfibrils of bacterial cellulose, produced by a process
5 which comprises the following steps:

 culturing a cellulose-producing
microorganism, capable of reversing its direction
during cellulose synthesis, in a nutrient medium
10 comprising an agent which interferes with
crystallization, but not polymerization, of said
cellulose, wherein said medium is contained in
and said culturing occurs in an enclosed plastic
container;

15 withdrawing said cellulose produced from
said culture; and

 forming said cellulose into an article.

20 2. The article as claimed in Claim 1, wherein
said microorganism is selected from the genera
Acetobacter, Rhizobium, Agrobacterium, Pseudomonas, or
Alcaligenes.

25 3. An article as claimed in Claim 2, wherein
said microorganism is selected from the genus Acetobacter.

30 4. An article as claimed in Claim 3, wherein
said microorganism is Acetobacter xylinum.

35 5. An article as claimed in Claim 4, wherein
said microorganism is the NQ-5 strain (ATCC 53582) of
Acetobacter xylinum.

1 6. An article as claimed in Claim 1, wherein
said agent is selected from glycerol, polyethylene glycol
or carboxmethylcellulose.

5 7. An article as claimed in Claim 6, wherein
said agent is carboxymethylcellulose..

10 8. An article as claimed in Claim 1, comprising
the further step of grafting polyacrylonitrile onto said
cellulose.

 9. An article as claimed in Claim 1, which is
formed into a sheet.

15 10. An article as claimed in Claim 9, wherein
said sheet is paper.

 11. An article as claimed in Claim 1, which
further comprises magnetic material.

20 12. An article as claimed in Claim 1, which
further comprises an electrical material.

25 13. An article as claimed in Claim 10, wherein
said process comprises the further steps of dyeing select
cellulose fibers and forming said paper into currency.

 14. An article as claimed in Claim 1, which
further comprises a thermosetting resin.

30 15. An article as claimed in Claim 1, wherein
said cellulose is formed into a film of a thickness of
less than about 0.1 micron.

1 16. An article as claimed in Claim 1, comprising
the further steps of forming said cellulose into a film
and vapor depositing an inorganic material onto said
cellulose film.

5 17. An article as claimed in Claim 1, comprising
the further steps of forming said cellulose into a film
and epitaxially growing an inorganic material on said
cellulose.

10 18. An article as claimed in Claim 1, which is
formed into a cloth shape.

15 19. An article as claimed in Claim 1, wherein
said process comprises the further step of freeze-drying
said cellulose.

20 20. An article as claimed in Claim 1, which is
formed into a foodstuff.

25 21. A process for producing an article of
manufacture from bacterial cellulose, comprising the steps
of:

25 culturing a cellulose-producing
microorganism, capable of reversing its direction
during cellulose synthesis, in a nutrient medium
comprising an agent which interferes with
crystallization, but not polymerization, of said
30 cellulose, wherein said medium is contained in
and said culturing occurs in an enclosed plastic
container;

35 withdrawing said cellulose produced from
said culture; and

forming said cellulose into an article.

INTERNATIONAL SEARCH REPORT

International Application No.

PCT/US89/02355

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) ⁸ According to International Patent Classification (IPC) or to both National Classification and IPC IPC (4): C 12 P 19/04; C 12 R 1/01, 1/02, 1/05, 1/38, 1/41 U. S. Cl: 435/101,822,823,829,874,878; 536/30,56																										
II. FIELDS SEARCHED <div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black;">Minimum Documentation Searched ⁷</div> <table style="width: 100%; border-collapse: collapse;"> <tr> <th style="width: 20%; border: 1px solid black; text-align: left;">Classification System</th> <th style="border: 1px solid black; text-align: left;">Classification Symbols</th> </tr> <tr> <td style="border: 1px solid black; text-align: center; vertical-align: middle;">U. S.</td> <td style="border: 1px solid black;">435/101,822,823,829,874,878; 536/30,56</td> </tr> </table> <div style="text-align: center; border-top: 1px solid black; border-bottom: 1px solid black;">Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched ⁸</div>			Classification System	Classification Symbols	U. S.	435/101,822,823,829,874,878; 536/30,56																				
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III. DOCUMENTS CONSIDERED TO BE RELEVANT ⁹ <table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th style="width: 10%; border: 1px solid black;">Category [*]</th> <th style="border: 1px solid black;">Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²</th> <th style="border: 1px solid black;">Relevant to Claim No. ¹³</th> </tr> </thead> <tbody> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td>US, A, 4,378,431 (Brown, Jr.) 29 March 1982 See the entire document.</td> <td style="vertical-align: top;">1-12 and 14-21</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y, P</td> <td>US, A, 4,788,146 (Ring et al.) 29 Nov. 1988 See the entire document.</td> <td style="vertical-align: top;">1-12 and 14-21</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">X</td> <td>Dillingham et al. (1961), <u>Bacterial Proceedings</u>, Abstract No. A68 See the entire document.</td> <td style="vertical-align: top;">1 and 3-4</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td>Dillingham et al. (1961), <u>Bacterial Proceedings</u>, Abstract No. A68 See the entire document.</td> <td style="vertical-align: top;">1-4,6-7 and 17-18</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td>US, A, 4,400,466 (Azoulay) 23 August 1983 See abstract and columns 1-6.</td> <td style="vertical-align: top;">1-4</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td>US, A, 4,692,408 (Banks et al.) 08 Sept. 1987 See abstract and columns 1-6.</td> <td style="vertical-align: top;">1-4</td> </tr> <tr> <td style="text-align: center; vertical-align: top;">Y</td> <td>US, A, 4,352,882 (Maury) 05 October 1982 See the entire document.</td> <td style="vertical-align: top;">1-4</td> </tr> </tbody> </table>			Category [*]	Citation of Document, ¹¹ with indication, where appropriate, of the relevant passages ¹²	Relevant to Claim No. ¹³	Y	US, A, 4,378,431 (Brown, Jr.) 29 March 1982 See the entire document.	1-12 and 14-21	Y, P	US, A, 4,788,146 (Ring et al.) 29 Nov. 1988 See the entire document.	1-12 and 14-21	X	Dillingham et al. (1961), <u>Bacterial Proceedings</u> , Abstract No. A68 See the entire document.	1 and 3-4	Y	Dillingham et al. (1961), <u>Bacterial Proceedings</u> , Abstract No. A68 See the entire document.	1-4,6-7 and 17-18	Y	US, A, 4,400,466 (Azoulay) 23 August 1983 See abstract and columns 1-6.	1-4	Y	US, A, 4,692,408 (Banks et al.) 08 Sept. 1987 See abstract and columns 1-6.	1-4	Y	US, A, 4,352,882 (Maury) 05 October 1982 See the entire document.	1-4
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<div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <p>[*] Special categories of cited documents: ¹⁰</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"Δ" document member of the same patent family</p> </div> </div>																										
IV. CERTIFICATION <table style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%; border: 1px solid black; vertical-align: top;"> Date of the Actual Completion of the International Search <div style="font-size: 1.2em; font-family: cursive;">07 September 1989</div> </td> <td style="width: 50%; border: 1px solid black; vertical-align: top;"> Date of Mailing of this International Search Report <div style="font-size: 1.2em; font-family: cursive;">24 OCT 1989</div> </td> </tr> <tr> <td style="border: 1px solid black; vertical-align: top;"> International Searching Authority <div style="text-align: center; font-weight: bold;">ISA/US</div> </td> <td style="border: 1px solid black; vertical-align: top;"> Signature of Authorized Officer <div style="font-family: cursive; font-size: 1.1em;">Pamela S. Webber</div> <div style="text-align: center;">Pamela S. Webber</div> </td> </tr> </table>			Date of the Actual Completion of the International Search <div style="font-size: 1.2em; font-family: cursive;">07 September 1989</div>	Date of Mailing of this International Search Report <div style="font-size: 1.2em; font-family: cursive;">24 OCT 1989</div>	International Searching Authority <div style="text-align: center; font-weight: bold;">ISA/US</div>	Signature of Authorized Officer <div style="font-family: cursive; font-size: 1.1em;">Pamela S. Webber</div> <div style="text-align: center;">Pamela S. Webber</div>																				
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FURTHER INFORMATION CONTINUED FROM THE SECOND SHEET		
Y	GB, B, 2,065,688 (Griffith et al.) 05 October 1982 See the entire document.	1-4
Y	US, A, 4,745,058 (Townesley) 17 May 1988 See the entire document.	6 and 7
Y	US, A, 4,416,993 (McKeown) 22 Nov. 1983 See abstract and columns 1-4.	6 and 7
Y	JOURNAL OF APPLIED POLYMER SCIENCE: APPLIED POLYMER SYMPOSIUM, vol. 37, 1983, pages 33-78, Brown, Jr. et. al. See the entire document.	1-21
V. <input type="checkbox"/> OBSERVATIONS WHERE CERTAIN CLAIMS WERE FOUND UNSEARCHABLE¹		
This international search report has not been established in respect of certain claims under Article 17(2) (a) for the following reasons:		
1. <input type="checkbox"/> Claim numbers _____, because they relate to subject matter ¹² not required to be searched by this Authority, namely:		
2. <input type="checkbox"/> Claim numbers _____, because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out ¹³ , specifically:		
3. <input type="checkbox"/> Claim numbers _____, because they are dependent claims not drafted in accordance with the second and third sentences of PCT Rule 6.4(a).		
VI. <input type="checkbox"/> OBSERVATIONS WHERE UNITY OF INVENTION IS LACKING²		
This International Searching Authority found multiple inventions in this international application as follows:		
1. <input type="checkbox"/> As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims of the international application.		
2. <input type="checkbox"/> As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims of the international application for which fees were paid, specifically claims:		
3. <input type="checkbox"/> No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claim numbers:		
4. <input type="checkbox"/> As all searchable claims could be searched without effort justifying an additional fee, the International Searching Authority did not invite payment of any additional fee.		
Remark on Protest		
<input type="checkbox"/> The additional search fees were accompanied by applicant's protest.		
<input type="checkbox"/> No protest accompanied the payment of additional search fees.		

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)		
Category *	Citation of Document, with indication, where appropriate, of the relevant passages	Relevant to Claim No
Y	"Cellulose chemistry and its applications," Editors T. P. Nevell et al., 1985, Ellis Horwood Ltd, New York, US; See entire document.	1-21
Y,L	SCIENCE, vol. 218, 1982, The American Association for the Advancement of Science; R. Malcolm Brown, Jr. et al.: "The Experimental Induction of Altered Nonmicrofibrillar Cellulose"	1-21
Y	JOURNAL OF CELL BIOLOGY, vol. 94, 1982, pages 64-69, The Rockefeller University Press; Haigler et al.: "Alteration of In Vivo Cellulose Ribbon Assembly by Carboxymethylcellulose and Other Cellulose Derivatives.	1-21
A	INTERNATIONAL SYMPOSIUM ON WOOD AND PULPING CHEMISTRY, June 9-12, 1981; vol. 3, R. Malcolm Brown, Jr. "Integration of Biochemical and Visual Approaches to the Study of Cellulose Biosynthesis and Degradation."	1-21